ELECTRONIC RESEARCH DIVISION

CLEVITE CORPORATION

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CLEVELAND, OHIO

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ABSTRACT

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This First Quarterly Report discusses the work performed under
Contract NAS3-2795 during the period October 1, 1963 through December 31,
1963.   This is a continuation of work performed under contract NAS7-293.

The effort expended on CdS solar cells during this quarter has been
devoted to obtaining reproducible films and cells, and to fabricating CdS
cells on duPont H-Film.   Fifty two evaporations were completed, yielding
217 films on glass, 50 films on duPont H-Film, two films on copper foil
and one attempt on a polystyrene substrate.   Improvements in the evapora-
tion of films and in film processing resulted in "standard" films with quite
uniform electrical properties.   Standard cells permit observation of a
number of trends, notably the dependence of cell quality on evaporation
rate and film thickness.   Investigation of the problem of cell deterioration
has begun, and data are presented to show that water vapor is not the only
agent responsible.   Experiments are described whose aim is to identify
these other agents.   Current-voltage characteristics typical of cells fabri-
cated in this laboratory are presented and discussed.   Mathematical models
of solar cells are described and their possible extension to CdS are discussed.

The "wet chemistry" approach has led to the room temperature
deposition of CdS mirrors on glass.   This technique is described.

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LIST OF ILLUSTRATIONS

STUDY OF THIN FILM LARGE AREA
PHOTOVOLTAIC SOLAR ENERGY CONVERTER

First Quarterly Report
October 1, 1963 - December 31, 1963
Contract No. NAS3-2795

1. INTRODUCTION

Present day photovoltaic solar energy converters are made of many small silicon cells, each of which must be soldered into a series-parallel array to provide the required electrical power for a given satellite system. A typical silicon cell is approximately 1 cm x 2 cm x 0.05 cm in size and has an efficiency of ten or twelve percent; therefore, each cell delivers twenty or thirty milliwatts of power and many cells are required to power a satellite. Furthermore, each cell is very much thicker than its p-n junction; consequently, unnecessary weight is involved in a silicon solar cell array.

It is the purpose of this contract to develop a large area thin film photovoltaic solar energy converter. It is hoped eventually that cadmium sulfide cells one foot square and one or two mils thick will be produced, although for the present smaller areas will be studied. The highest efficiency of presently available thin film cadmium sulfide cells is around three percent over relatively small areas, and although the density of cadmium sulfide is twice that of silicon, the thickness of a cadmium sulfide cell is only about one-tenth that of a silicon cell. Thus, large area cadmium sulfide cells would compare favorably on a weight-per-watt and cost-per-watt basis with silicon cells. Furthermore, cadmium sulfide film cells have been produced on flexible metal and high-temperature organic plastic substrates, representing a great potential improvement over the rigid backing required by single crystal silicon cells. There is a growing body of evidence that cadmium sulfide cells are more resistant to radiation than silicon cells.

This report covers work done during the first quarter of this contract.

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2. EVAPORATED FILM CELLS

Fifty-two evaporations were completed during this quarter, yielding 217 films on glass, 50 films on H-Film, two films on copper foil and one attempt on a polystyrene substrate, for a total of 270 films. These are evaporations 65 through 116.

Most of the films deposited on glass were standard films, and most of these were processed into photovoltaic cells. The evaporations on H-Film established the parameters necessary to produce cells with good conversion efficiencies, i. e., in the 1.5 to 2.5 percent range. The films on copper foil substrates did not develop a photovoltage and efforts in this direction are suspended. The evaporation on the polystyrene substrate ended when the substrate melted.

Details concerning the various cells and evaporation experiments are given in Sections 3 and 4.

2.1 Evaporator

The quartz evaporation crucibles have been modified to allow insertion of a quartz shielded thermocouple into the evaporant. This permits direct monitoring of the evaporator temperature and closer control over the evaporated films. The maximum evaporation temperature during a good run is about 1200°C. A time-temperature profile reveals that the evaporation temperature reaches a maximum and then drops quickly. The temperature drop appears to occur when the charge has evaporated sufficiently to uncover the thermojunction. This stage is reached in about two minutes, which is usually sufficient for the required film deposit.

A change in maximum evaporation temperature is an indication of a change in the temperature profile along the evaporation crucible. This in turn permits an assessment of the further usefulness of the heater coil. If, after two or three evaporations, the heater coil sags, a change in the temperature profile is observed. The heater coil must then be replaced.

A number of heater coil configurations have been employed to try to increase the useful life of the coil. The heater coil is a 3/4 inch diameter

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helix of tantalum wire and is mounted vertically. Its top five turns are spaced 1/32 inch apart and the remaining ten turns are spaced 3/32 inch apart. This is the best configuration for obtaining good films. After a few evaporations the coil tends to sag under its own weight, resulting in a change in the turns spacing. This leads to films not suitable for good photovoltaic cells. Attempts to retain the shape of the coil by packing it with a refractory fiber have not been very successful. Other attempts, including a grooved boron nitride cylinder and packing with quartz cloth were partially successful: the boron nitride is too good a thermal conductor, and the quartz cloth is not rigid enough.

It is planned to replace the tantalum coil with a tungsten coil which will be stronger and to modify the coil mount so that changing coils will be a relatively simple task. Both changes are in process and should be completed within a week or two. Inquiries are being made to locate a grooved quartz cylinder to hold the heater coil.

2.2 Substrate

In an attempt to overcome the tendency of H-Film to curl after being coated with CdS, an embossed film has been obtained and will be used as a substrate in the near future. The pattern embossed on the H-Film is an array of closely packed hemispheres. The embossed pieces are much more rigid than the plain pieces. CdS will be evaporated onto the concave side of the pattern with the expectation that the concavity will, at most, be flattened out.

3. PROCESSING OF FILMS

The efficiencies of standard glass backed cells are averaging approximately 1.8 percent. Only a few films fall outside the range of 1.5 to 2.5 percent. Those that fall below this range are always found to have some obvious deficiency such as abnormally high resistance or an imperfectly formed barrier layer. On the other hand, a few cells are always found with efficiencies significantly above this range indicating that the average cell obtained from the standard process is not yet the optimum cell. For example,

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although the average efficiency appears to be around two percent, a significant number of cells is found to have efficiencies in slight excess of three percent.

The average efficiency level fell to a nominal one percent, for a time, when a newly formulated CdS charge, boule T-16, was put into use. This drop was traced to a deficiency in indium doping caused, apparently, by control failure in the sintering operation. (Although powder charges only are evaporated, the powders are obtained by grinding sintered boules. The powdered charge is again sintered in place in the evaporation crucible.) A new boule, T-17, with approximately the correct indium concentration has been obtained and the donor concentration in the evaporated films is being corrected by adjusting the ratio of doped to undoped material in the charge. Evaporations 93 through 99 were affected by the weakly doped boule T-16; evaporation 100H (H following the evaporation number denotes an H-Film substrate) employed a combination of boules T-10 and T-11; and evaporations 101 through 103 employed a combination of boules T-15 and T-17 to obtain good films.

The necessary frames, masks and other hardware required for evaporations on H-Film have been obtained and successfully employed to evaporate independent CdS films on a common H-Film substrate. A square array of 9 films, each about 7/8 inch square, is evaporated on a single 4 inch x 4 inch H-Film substrate. The individual films are processed into photovoltaic cells and may be connected in series or parallel or in combination to form photovoltaic batteries. The best such array obtained so far (actually obtained a few days after the end of this quarter) has 8 individual cells with efficiencies between 1. 9 and 2. 6 percent and one cell of 1. 5 percent.

Large area cells on H-Film have generally shown one percent efficiency over an area of 35 to 40 cm^2. Adhesion of CdS to H-Film is good, and is greatly improved when the H-Film surface is roughened by light sandblasting.

When a sufficient number of H-Film cells have been processed to establish the processing parameters, glass substrate cells will be made

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only for quality control and special purposes. H-Film cells are heat treated in a forced convection oven at 250°C instead of on the 300°C hot plate generally employed for glass backed cells.

Table 3-1 summarizes the photovoltaic cells processed during this quarter. Generally, three cells are listed under each evaporation heading: one listing showing the highest efficiency cell obtained, one listing showing the lowest efficiency cell obtained, and one listing showing the properties derivable from the Hall effect and resistivity measurements. Not every cell from a given evaporation was processed. Needless to say, the high efficiency listing shows the least quality to be expected from a given evaporation; the low efficiency listing is generally attributable to some departure from the standard process. It should be noted, however, that even the high efficiency listing may mean a departure from the standard process.

Column 1 of Table 3-1 lists the cell number, the first part of which denotes the evaporation number and the last part of which denotes the particular film from which the cell was made. If the substrate was H-Film, the evaporation number is followed by the letter H. The second and fourth columns give the open circuit voltage, V_{oc}, and output voltage at the maximum power transfer point, V_m, respectively, in volts; the third and fifth columns give the short circuit current density, I_{sc}, and output current density at the maximum power transfer point, I_m, respectively, in milliamperes per square centimeter; the sixth column gives the active area of the cell (area of the barrier layer) in cm^2; the seventh column gives the conversion efficiency of the cell at its maximum power transfer point, measured in natural sunlight or in artificial light equivalent to $100\ mW\text{-}cm^{-2}$ of natural sunlight; the eighth column gives the fill factor which is defined as the ratio of the product $V_m I_m$ to the product $V_{oc} I_{sc}$; the ninth column gives the average thickness of the unprocessed CdS film in cm; the tenth column gives the dark resistivity, ρ, of the CdS film in ohm cm; and the last column gives the charge carrier concentration, n, in cm^{-3} determined from the Hall coefficient of the film, measured in the dark.

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TABLE 3-1. Properties of CdS Film Cells

Cell No.	V_{oc}, volts	I_{sc}, mA-cm^{-2}	V_m, volts	I_m, mA-cm^{-2}	Active Area, cm^2	Efficiency, percent	Fill Factor	Thickness, cm	Resistivity, ohm-cm	Carrier Concentration, n, cm^{-3}
65-6A	0.50	12.4	0.38	9.0	0.61	3.4	0.55	3.0×10^{-3}	0.073	1.1×10^{19}
65-2	0.45	3.0	0.30	2.0	3.8	0.6	0.45			
66-1	Copper foil substrate. No photovoltage.									
67-1	Copper foil substrate. No photovoltage.									
68-6	0.46	6.3	0.32	3.9	0.12	1.5	0.43			
68-8	0.44	4.6	0.30	3.2	8.2	1.0	0.47			
68-7								4.25×10^{-3}	0.054	9.9×10^{18}
69-7	0.52	8.0	0.40	6.2	7.14	2.5	0.60			
69-2	0.42	2.2	0.30	1.4	7.74	0.4	0.46			
69-3								4.6×10^{-3}	0.035	8.7×10^{18}
70-4	0.41	6.6	0.27	4.6	6.72	1.2	0.46			
70-3	0.44	4.0	0.30	2.6	2.30	0.1	0.45			
70-2								3.0×10^{-3}	0.053	7.4×10^{18}
71-2	0.44	3.8	0.30	2.5	7.19	0.7	0.44			
71-7	0.43	2.1	0.27	1.4	3.50	0.4	0.43			
71-4								3.5×10^{-3}	0.093	4.8×10^{18}
72-6	0.44	4.2	0.32	3.2	7.95	1.0	0.55			
72-7	0.44	2.4	0.34	1.8	7.83	0.6	0.56			
72-1								2.5×10^{-3}	0.19	4.5×10^{18}
73-7	0.50	5.1	0.33	3.5	6.40	1.2	0.45			
73-6	0.48	2.6	0.34	2.1	8.40	0.7	0.58			
73-1								2.0×10^{-3}	0.31	4.7×10^{18}
74H-1	H-Film substrate. No adherence.									
75-1	Polystyrene substrate. Melted.									
76H-1	H-Film substrate. No adherence.									
77H-1	0.36	1.9	0.22	1.1	2.48	0.3	0.35			

TABLE 3-1. Properties of CdS Film Cells (Cont'd)

Cell No.	V_{oc}, volts	I_{sc}, mA-cm^{-2}	V_m, volts	I_m, mA-cm^{-2}	Active Area, cm^2	Efficiency, percent	Fill Factor	Thickness, cm	Resistivity, ohm-cm	Carrier Concentration, n, cm^{-3}
78H-1	H-Film substrate. Curled and flaked.									
79-1	0.47	4.1	0.37	3.4	7.53	1.3	0.67			
79-8	0.48	3.0	0.35	2.3	6.60	0.8	0.57			
79-2								1.8×10^{-3}	0.17	7.8×10^{18}
80H-1	0.45	2.9	0.25	1.6	39.4	0.4	0.31			
81-1	0.47	1.6	0.34	1.3	60.6	0.4	0.58			
82-1	0.47	10.7	0.33	8.3	7.4	2.7	0.54			
82-8	0.47	6.1	0.34	3.7	0.8	1.3	0.44			
82-3								6.1×10^{-3}	0.026	9.5×10^{18}
83-8	0.42	4.7	0.32	3.5	8.60	1.1	0.57			
83-6	0.42	3.5	0.30	2.3	8.00	0.7	0.47			
83-1								5.9×10^{-3}	0.054	6.6×10^{18}
84H-1	0.46	2.9	0.35	2.5	21.6	0.9	0.64			
85-8								4.9×10^{-3}	0.030	8.6×10^{18}
86-1	0.51	8.3	0.38	5.8	5.9	2.2	0.52			
86-7	0.47	2.0	0.34	1.3	7.2	0.5	0.47			
86-6								5.6×10^{-3}	0.024	9.6×10^{18}
87-1	0.51	9.8	0.37	8.0	7.0	2.9	0.60			
87-6	0.46	4.3	0.35	3.0	8.0	1.1	0.54			
87-2								4.2×10^{-3}	0.047	7.7×10^{18}
88-1	0.49	3.3	0.39	2.9	7.8	1.2	0.41			
88-8	0.45	3.7	0.34	2.9	7.3	1.0	0.60			
88-2								3.4×10^{-3}	0.085	7.1×10^{18}

TABLE 3-1. Properties of CdS Film Cells (Cont'd)

Cell No.	V_{oc}, volts	I_{sc}, mA-cm^{-2}	V_m, volts	I_m, mA-cm^{-2}	Active Area cm^2	Effi- ciency, percent	Fill Factor	Thick- ness, cm	Resist- ivity, ohm-cm	Carrier Concen- tration, n, cm^{-3}
89H-A	0.46	6.4	0.29	4.8	9.3	1.4	0.48			
89H-B	0.45	5.3	0.32	4.2	8.6	1.4	0.57			
89H-C	0.46	6.4	0.31	5.1	8.4	1.6	0.53			
89H-D	0.46	4.7	0.32	3.8	9.0	1.2	0.56			
89H-1*	0.45	5.1	0.30	3.8	35.3	1.1	0.49			
90H-1	0.45	3.8	0.31	2.8	34.8	0.9	0.51			
91H-A	0.44	4.8	0.31	3.5	9.3	1.2	0.55			
91H-B	0.44	3.8	0.33	2.9	9.3	0.9	0.53			
91H-C	0.43	5.9	0.30	4.6	9.9	1.4	0.54			
91H-D	0.44	5.7	0.30	4.4	9.0	1.3	0.53			
91H-1*	0.43	4.7	0.27	3.5	37.6	0.9	0.47			
92-4	0.45	8.9	0.31	6.7	8.4	2.1	0.52			
92-7	0.37	5.5	0.25	4.0	8.1	1.0	0.49			
92-8								4.3×10^{-3}	0.067	7.4×10^{18}
93-6	0.39	9.1	0.23	5.7	8.7	1.3	0.37			
93-3	0.43	3.1	0.32	2.6	8.0	0.8	0.63			
93-5								3.3×10^{-3}	0.29	5.5×10^{18}
94-4	0.41	9.2	0.27	6.4	8.4	1.7	0.46			
94-2	0.51	2.8	0.39	2.6	7.1	1.0	0.72			
94-1								3.1×10^{-3}	0.11	5.7×10^{18}
95H-1	0.45	1.1	0.30	0.9	44.2	0.3	0.54			
96-3	0.48	2.6	0.38	2.3	8.6	0.9	0.70			
96-2								3.2×10^{-3}	0.079	6.0×10^{18}
97H-1	H-Film substrate. High resistance because of Boule T-16.									
98-2	0.48	2.8	0.38	2.5	7.7	1.0	0.71			
98-3								3.3×10^{-3}	0.129	6.5×10^{18}

*Cells A, B, C, and D connected in parallel. Common CdS layer and separate barrier layers.

TABLE 3-1. Properties of CdS Film Cells (Cont'd)

Cell No.	V_{oc}, volts	I_{sc}, mA-cm^{-2}	V_m, volts	I_m, mA-cm^{-2}	Active Area, cm^2	Efficiency, percent	Fill Factor	Thickness, cm	Resistivity, ohm-cm	Carrier Concentration, n, cm^{-3}
99H-A	0.43	1.2	0.31	0.9	9.0	0.3	0.55			
99H-B	0.46	1.8	0.37	1.5	9.3	0.6	0.66			
99H-C	0.46	1.7	0.35	1.7	9.0	0.5	0.64			
99H-D	0.46	2.5	0.36	2.1	8.4	0.8	0.67			
99H-1*	0.40	1.2	0.36	0.9	35.7	0.3	0.49			
100H-1	Not processed.									
101-7	0.48	5.5	0.34	4.4	7.6	1.6	0.73			
101-4	0.27	4.1	0.16	2.6	8.4	0.4	0.37			
101-2								2.3×10^{-3}	0.074	7.4×10^{18}
102-3	0.41	8.7	0.27	6.3	8.1	1.7	0.47			
102-6	0.43	1.5	0.33	1.1	9.1	0.4	0.56			
102-2								1.7×10^{-3}	0.052	8.3×10^{18}
103-5	0.26	9.1	0.21	5.1	0.9	1.1	0.45			
103-7	0.41	1.3	0.30	0.9	7.6	0.3	0.54			
103-8								4.4×10^{-3}	0.015	1.4×10^{19}
104-2	0.45	7.6	0.33	5.8	7.8	1.9	0.56			
104-4	0.40	3.4	0.29	2.3	2.8	0.7	0.50			
104-7								3.8×10^{-3}	0.039	8.0×10^{18}
105-4	0.43	3.3	0.29	2.5	7.9	0.7	0.52			
105-6								1.6×10^{-3}	0.45	4.2×10^{18}
106H-1	Not processed.									
107H-1	Not processed.									
108-2	Electrical properties only.							2.3×10^{-3}	0.85	9.3×10^{17}
109-8B	0.48	10.9	0.39	8.6	0.7	3.3	0.59			
109-8A	0.48	9.4	0.38	6.6	0.7	2.5	0.56			
109-2								5.8×10^{-3}	0.033	7.3×10^{18}

*Cells A, B, C, and D connected in parallel. Common CdS layer and separate barrier layers.

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TABLE 3-1. Properties of CdS Film Cells (Cont'd)

Cell No.	V_{oc}, volts	I_{sc}, mA-cm^{-2}	V_m, volts	I_m, mA-cm^{-2}	Active Area, cm^2	Efficiency, percent	Fill Factor	Thickness, cm	Resistivity, ohm-cm	Carrier Concentration, n, cm^{-3}
110-2	0.43	7.2	0.32	5.8	8.0	1.8	0.59			
110-7	0.32	3.2	0.22	2.1	7.8	0.5	0.44			
110-1								4.8×10^{-3}	0.021	9.1×10^{18}
111-6A	0.45	7.6	0.35	5.8	3.0	2.0	0.59			
111-6B	0.30	0.5	0.18	0.4	2.9	0.1	0.40			
111-3								4.8×10^{-3}	0.017	8.6×10^{18}
112-1C	0.39	8.0	0.25	5.5	3.2	1.3	0.43			
112-1B	0.40	3.2	0.20	1.8	3.8	0.4	0.29			
113H-1	Not processed.									
114H-1	Not processed.									
115-1	Not processed.									
116-3	Electrical properties only.							5.9×10^{-3}	0.022	7.7×10^{18}

4. MEASUREMENTS

Determinations of current-voltage characteristics of CdS cells are made on a routine basis, as are resistivity and charge carrier concentrations, on at least one film from each evaporation.

4.1 CdS Film Cell Deterioration

CdS film cell deterioration was mentioned briefly in the Fourth Quarterly Report.[1] Other workers have also reported on cell deterioration but generally disagree on the amount and nature of the effect. An attractive hypothesis is that cell deterioration is caused by water vapor attacking the barrier layer, and indeed, in at least one instance,[2] protective, vapor proof encapsulation of CdS thin film solar cells has been effective in eliminating deterioration. On the other hand, Chamberlin[3] reports that no deterioration was observed in six months on unprotected cells. It is evident from Fig. 4-1 that the situation is not so simple in the case of back wall cells fabricated in this laboratory. It has been repeatedly observed that most, but not all, cells left exposed to the laboratory ambient will rapidly decrease in efficiency by as much as 50 percent. To determine whether this was all due to water vapor in the barrier layer, a few cells were placed in a vacuum desiccator immediately after fabrication and obtaining an initial current-voltage characteristic. It was expected that the efficiency of such a cell might at first decrease but that after a few days would recover its initial value. Figure 4-1 shows that this does not happen. Instead, after an initial rapid decrease lasting 4 or 5 days, the rate of decrease is still significant, albeit a great deal smaller than the initial rate. This is exemplified by the curves for cells 110-8 and 116-2. In order to observe this more carefully, two cells, 111-1 and 111-2, were chosen for the following experiment. The cells were identically and simultaneously fabricated from films evaporated side by side on a glass substrate. One cell, 111-1, was placed in a "dry" box, the other in a vacuum desiccator. Cell 111-1 deteriorated at more than three times the rate of cell 111-2 for about 10 days (see Fig. 4-1). Then, cell 111-1 decreased its rate of deterioration while that of 111-2 remained nearly constant. The data are insufficient for accurate comparisons, but it does
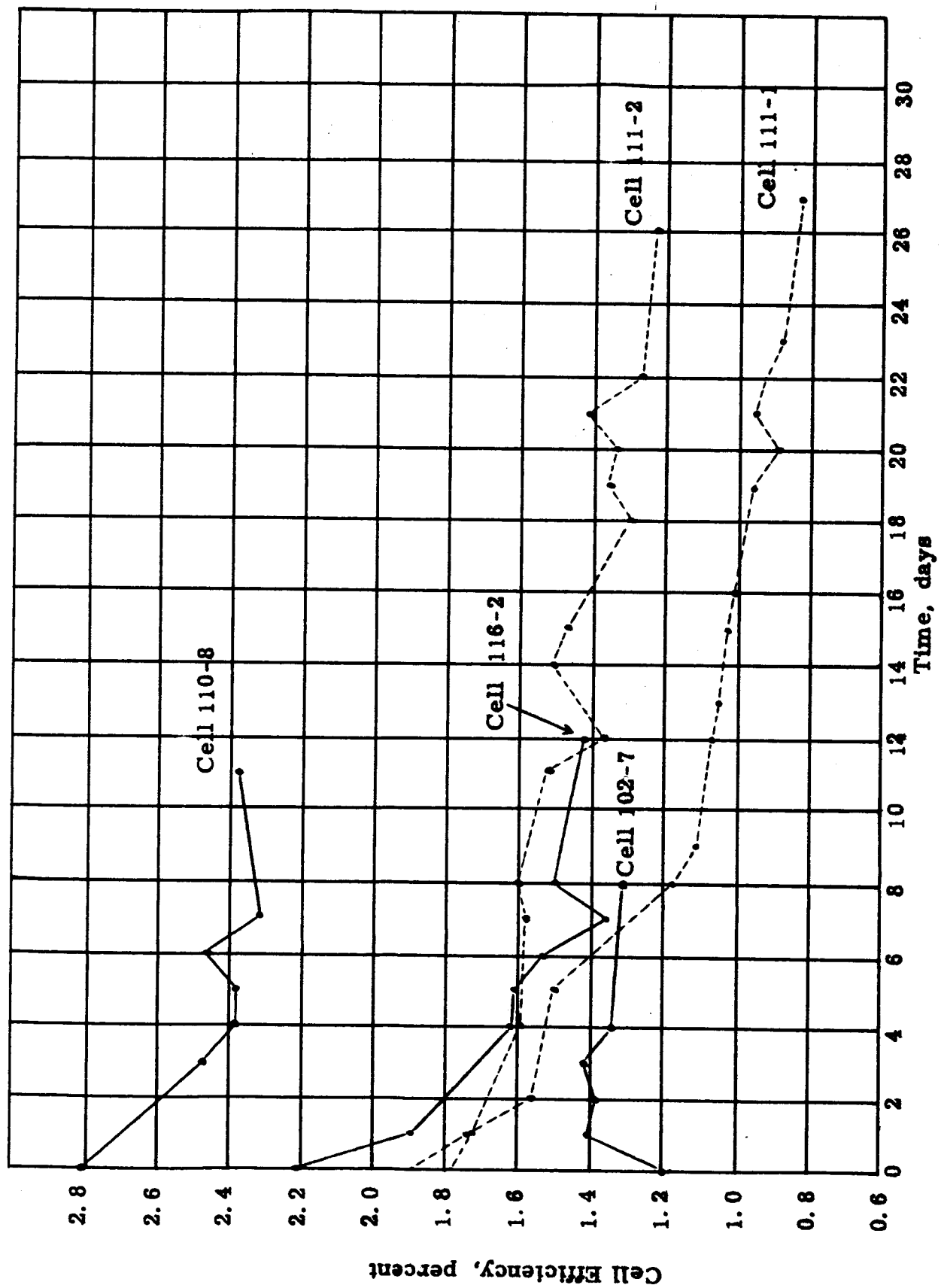
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FIGURE 4-1. DETERIORATION OF CdS FILM CELLS

constant. The data are insufficient for accurate comparisons, but it does seem as though the deterioration rate of cell 111-1 between 10 and 16 days is very nearly the same as that for cell 111-2. On the 16th day, cell 111-1 was placed in the vacuum desiccator with cell 111-2. This did not appear to have any effect on the deterioration rate of cell 111-1, for, after 27 days, the slope of 111-1 curve does not differ greatly from that of 111-2. Again, it must be emphasized that the data are insufficient for drawing any firm conclusions, but it appears that a dry atmosphere, while retarding the initial rate of deterioration, does not materially affect the long term rate of deterioration. Additional experiments are planned to determine whether a dry atmosphere actually is more effective in maintaining a higher efficiency level than the ambient atmosphere, as indicated by the curve of cell 110-8. One of these will involve heating a deteriorated cell in a vacuum or inert atmosphere to see if the efficiency recovers its initial value.

One might tentatively conclude that water vapor does cause an initial rapid deterioration of a film cell, which is not reversed by putting the cell into a completely dry atmosphere. One may also conclude that one or more other agencies are responsible, at least in part, for the continued deterioration observed in the completely dry atmosphere. Several possibilities come to mind. Among the most likely are donor migration into the barrier layer and impurities introduced with the slurry and not completely removed. To examine the donor migration possibility, a CdS film cell was fabricated without the usual soldered indium peripheral contact to the n-type layer. Instead, a peripheral contact of electroplated rhodium was formed and reinforced with conducting silver paint. Great care was taken while processing this cell to eliminate every possibility of indium contamination other than that deliberately introduced as a dopant during evaporation. The variation in efficiency with time for this cell (cell 102-7) is also shown in Fig. 4-1. An initial increase in efficiency is observed followed by a much more gradual deterioration of this cell than is observed for the indium electroded cells. This, then, suggests that the "impurities in the barrier layer" agency might also be at work. Experiments are planned to examine these possibilities in detail, as well as to continue the general deterioration study which will now

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become a major part of this program, particularly with reference to the H-Film substrate cells now being fabricated.

It should be mentioned that the first deterioration measurements were made on cell 88-5 and have continued for some 70 days. This cell was fabricated November 7, 1963 and had an initial efficiency of 2.2 percent. After 30 days it stabilized at about 1.5 percent. Its curve was not included in Fig. 4-1 because pressure contacts only can be applied to this cell and contact resistance caused a very wide scatter of experimental points. The cells described above all have terminal leads of silvered copper braid soldered to the peripheral electrode and cemented with conducting silver paint to the silver paint electrode covering the barrier layer. This effectively eliminates contact resistance. It should also be pointed out that the cells described by Fig. 4-1 (except for cell 111-1) would, most likely, all have deteriorated to less than 1 percent efficiency within a few days had they been left exposed to the ambient atmosphere.

4.2 Refractive Index and Reflecting Power of DuPont H-Film

An approximate value of the refractive index of duPont H-Film was obtained by measuring the apparent thickness with a short focus microscope. The measurement was made because the manufacturer's data notes only that the refractive index is in excess of 1.7. A small dot of ink was placed on each side of a piece of H-Film. A short focus microscope (magnification: 1140 X) was aligned perpendicular to the H-Film surface and focused on one of the dots. The microscope barrel was moved vertically to focus on the other dot and the displacement of the barrel measured with the microscope's vertical motion vernier. The average of 13 measurements was 0.00253 ± 0.00023 cm for the apparent distance between the two dots. The actual thickness of the H-Film (vertical distance between the dots) was measured with a micrometer caliper and found to be 0.00490 ± 0.00006 cm. The refractive index is the ratio of the real to the apparent thickness or

$$n = 1.94 \pm 0.18 \ . \tag{4-1}$$

Employing Fresnel's equation for reflecting power at a surface, it is possible to calculate the percentage loss of light incident on H-Film from air or vacuum. Fresnel's equation for normal incidence is

$$R = \left[\frac{(_1n_2 - 1)}{(_1n_2 + 1)} \right]^2 . \tag{4-2}$$

Thus for an air (or vacuum) interface, the reflection loss for light incident on H-Film from air is

$$R = \left[\frac{(1.94 - 1)}{(1.94 + 1)} \right]^2 = 0.0102 . \tag{4-3}$$

Thus only 1.02 percent of the incident light is lost by reflection. The reflection loss for the H-Film-CdS interface may also be calculated from Eq. (4-2) after calculating the relative refractive index for H-Film to CdS. The refractive index of CdS is about 2.5 (average), so the relative refractive index for light incident on CdS from H-Film is 2.5/1.94 = 1.29. Substituting this into Eq. (4-2),the reflection loss is found to be

$$R = \left[\frac{(1.29 - 1)}{(1.29 + 1)} \right]^2 = 0.016 . \tag{4-4}$$

Thus only 1.6 percent of the light is lost at the CdS-H-Film interface. This means that better than 97 percent of the incident light will enter the CdS layer of a solar cell.

4.3 Evaporation Rate, Cell Efficiency and Film Thickness

The data taken on cells from sixty evaporations were examined in various ways. One interesting result was obtained when the efficiency of the best cell from an evaporation was plotted against the deposition rate for that evaporation. These data are shown in Fig. 4-2, and it is obvious that the best cells are obtained for the highest deposition rates. Indeed, with but one exception, no cells with efficiency greater than two percent were obtained at an evaporation rate less than about 0.19 micron per second. If maximum

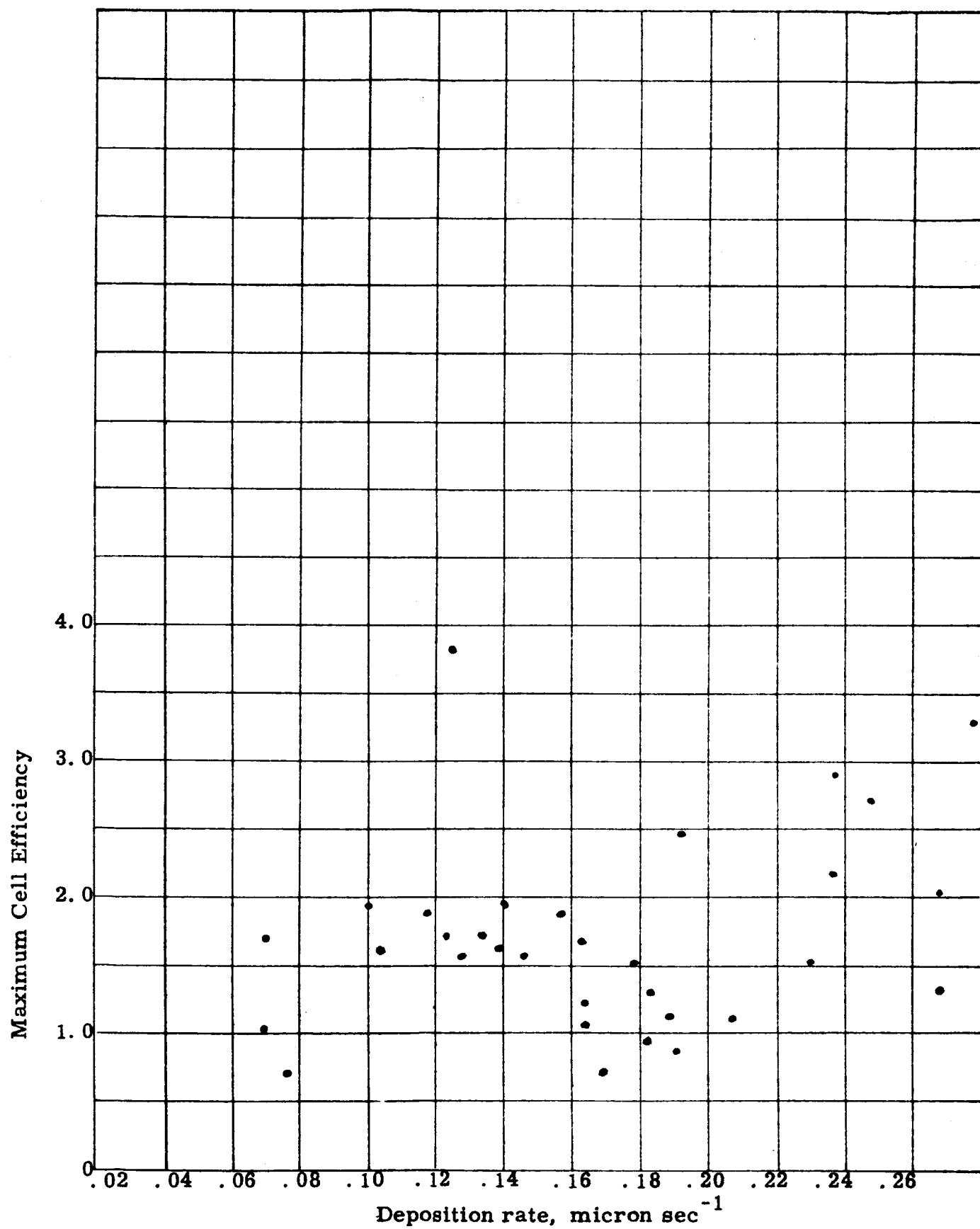CLEVITE CORPORATION   ELECTRONIC RESEARCH DIVISION

FIGURE 4-2.   EFFECT OF DEPOSITION RATE ON CELL EFFICIENCY

open circuit voltages and maximum short circuit currents are plotted in the same manner, it is observed that the maximum attainable current also increases with deposition rate, while no such trend is observed for open circuit voltages. On the other hand, if the same quantities are plotted against film thickness, it is immediately obvious that open circuit voltage, short circuit current and efficiency all increase with film thickness. The data for open circuit voltage and short circuit current are shown in Fig. 4-3.

4.4 Area Effect on Cell Efficiency

It has been hypothesized that cell efficiency might drop off as cell area increases, and further hypothesized that this might be corrected for by changing cell geometry from square to rectangular. To examine this effect, a mask was made with various sizes and shapes of holes so that a number of areas with various geometries could be evaporated simulataneously. This was done on a glass substrate and all cells processed identically. The results are given in Table 4-1. It appears at first glance that the hypotheses are correct, but the data are insufficient for detailed analysis. More experiments of this type will be performed.

TABLE 4-1. Area Effect on Cell Efficiency

Nominal Area	Efficiency
1/2 inch x 1/2 inch	0.89
3/4 inch x 3/4 inch	0.40
1 inch x 1 inch	0.57
2 inch x 2 inch	0.41
1/2 inch x 2 inch	1.34
1/2 inch x 3 1/2 inch	1..08
1 inch x 2 inch	0.89

4.5 Analysis of i-V Characteristics

Figures 4-4 through 4-8 are representative of the types of experimental i-V characteristics obtained from cells fabricated in this laboratory. The curves in Fig. 4-4 are typical of one percent cells, those in Fig. 4-5 are typical of two percent cells, and those in Fig. 4-6 of three percent cells.
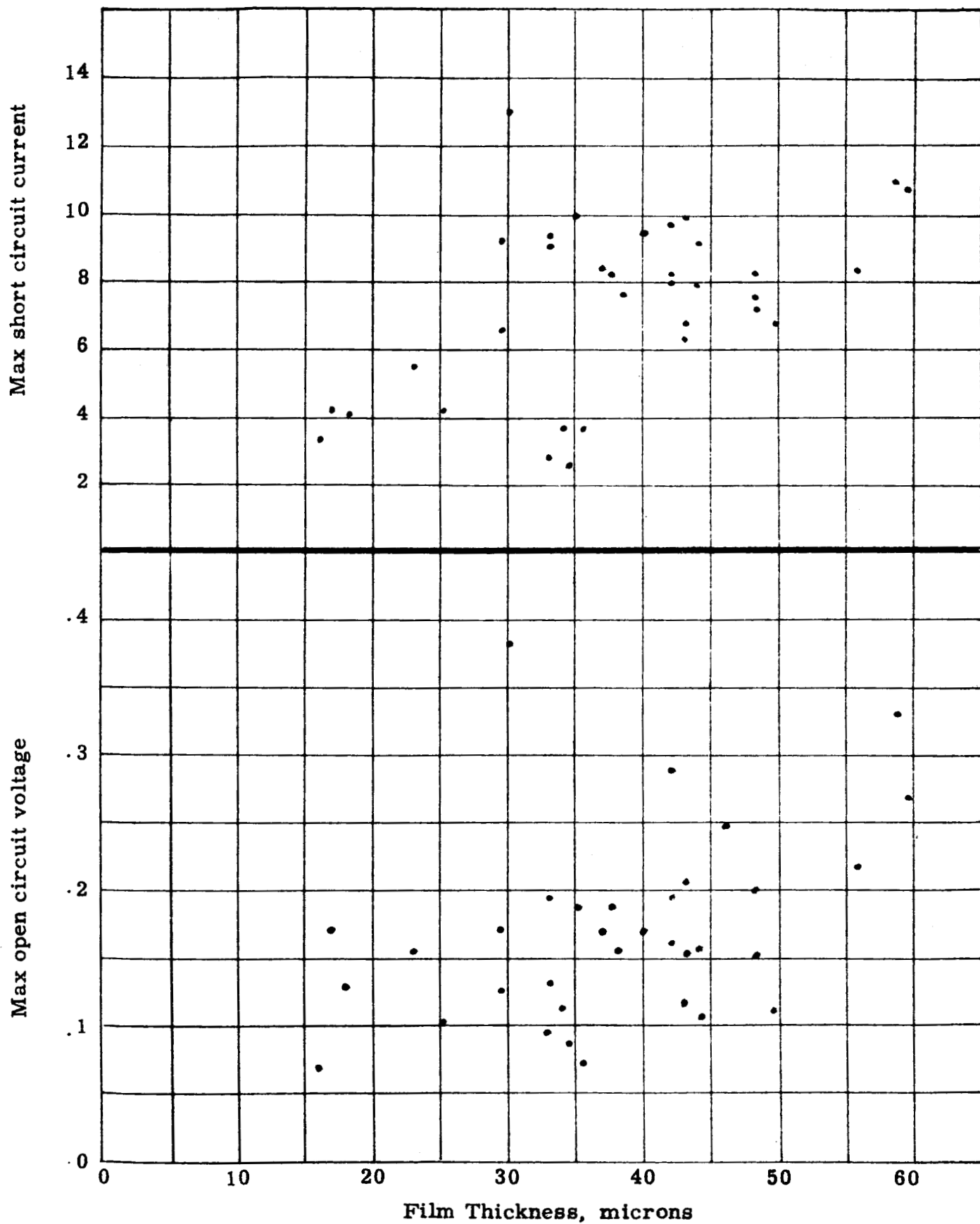
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FIGURE 4-3.   DEPENDENCY OF OPEN CIRCUIT VOLTAGE AND SHORT
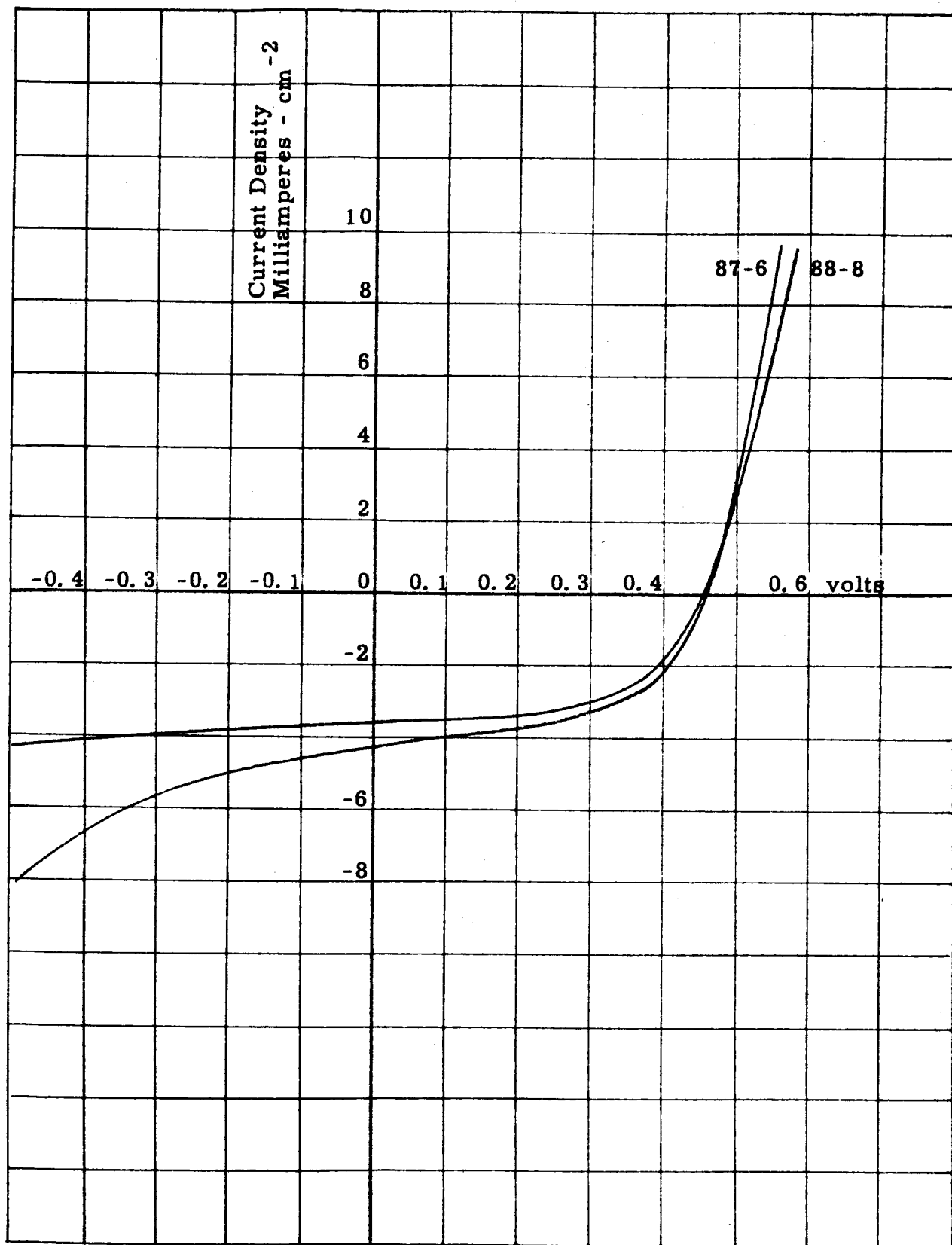CIRCUIT CURRENT ON FILM THICKNESS

FIGURE 4-4. CURRENT-VOLTAGE CHARACTERISTIC OF
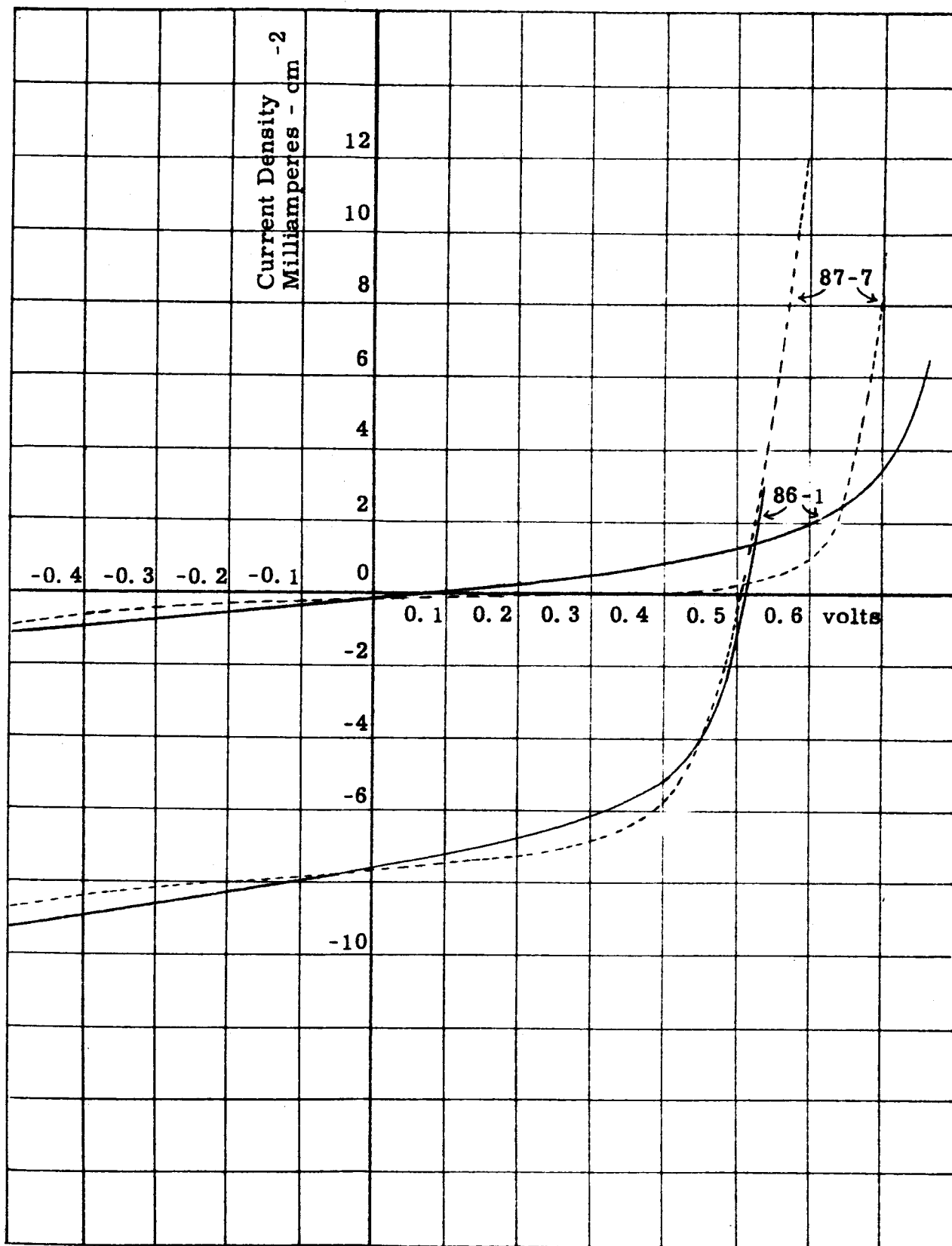CdS FILM CELLS 87-6 AND 88-7

FIGURE 4-5. CURRENT-VOLTAGE CHARACTERISTIC OF
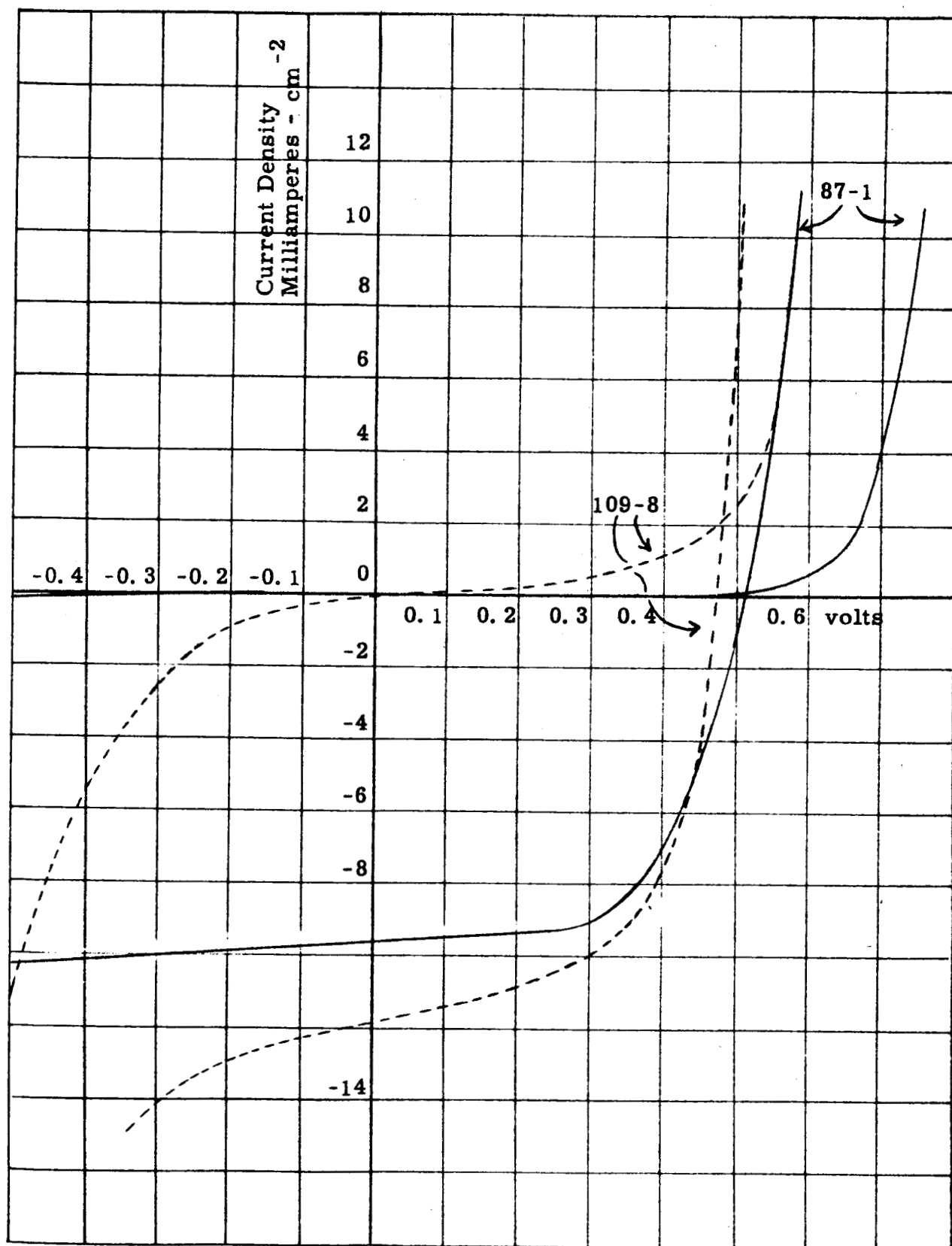CdS FILM CELLS 86-1 AND 87-7

FIGURE 4-6.   CURRENT-VOLTAGE CHARACTERISTIC OF
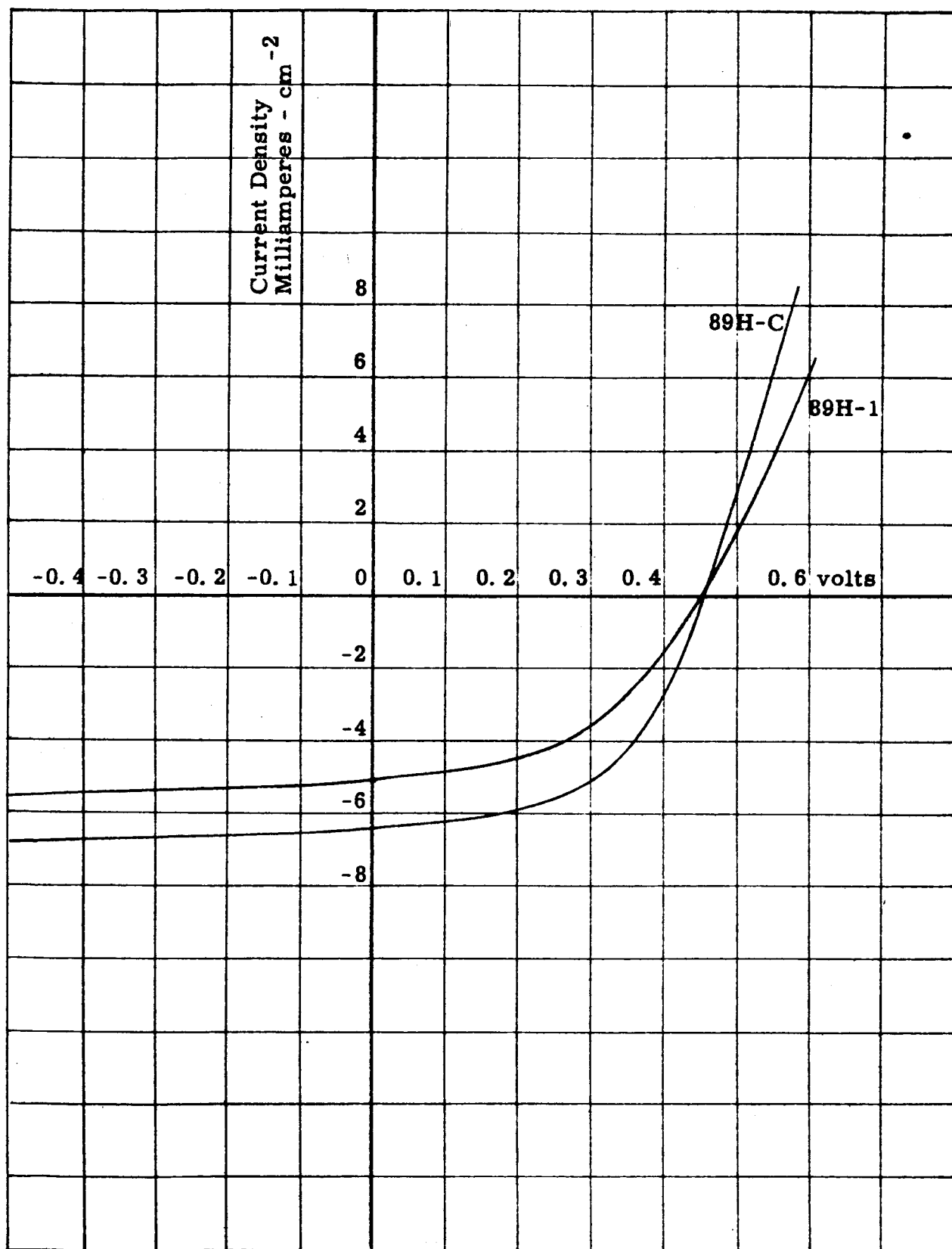CdS FILM CELLS 87-1 AND 109-8

FIGURE 4-7.   CURRENT-VOLTAGE CHARACTERISTIC OF
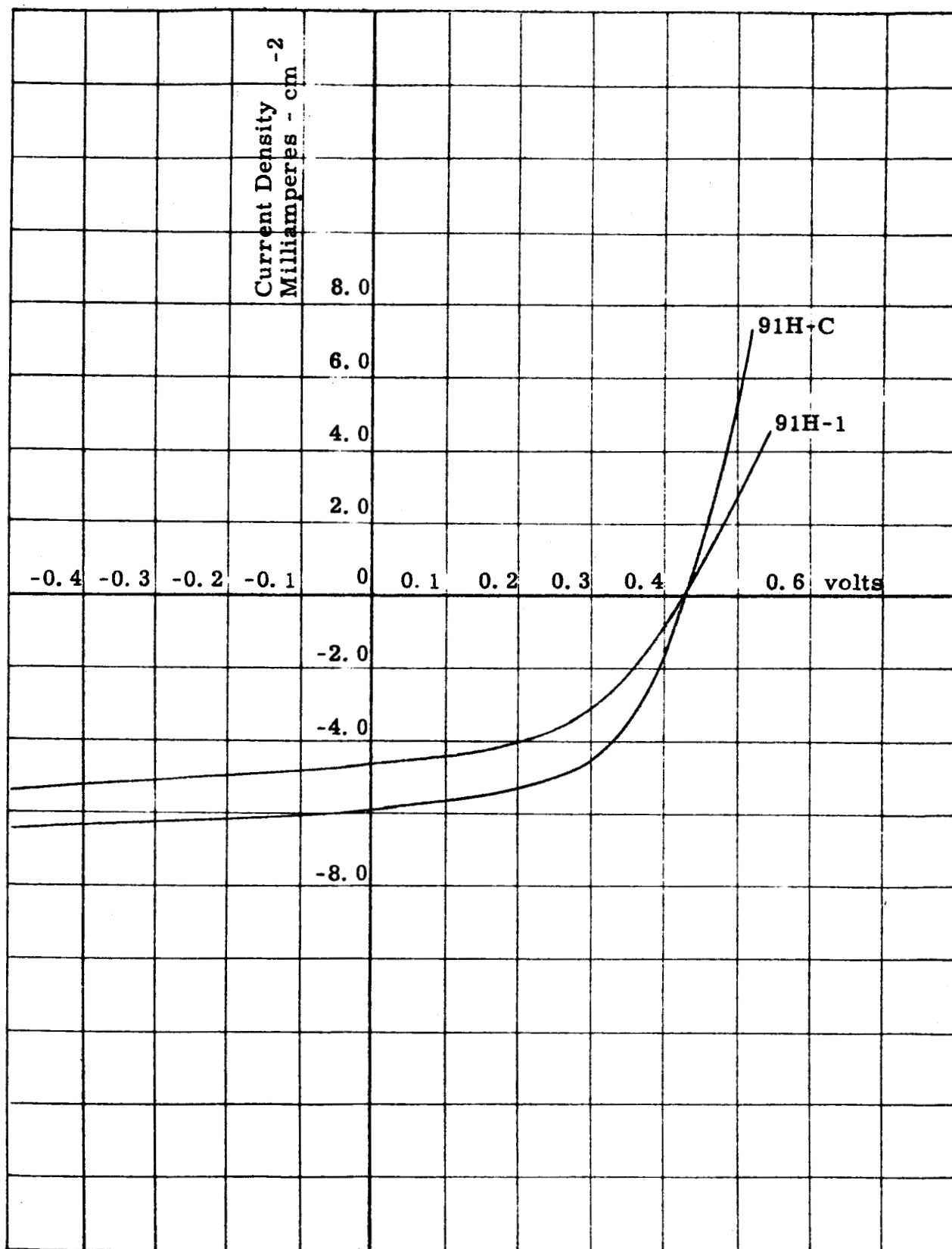CdS FILM CELL 89H-1.

FIGURE 4-8. CURRENT-VOLTAGE CHARACTERISTIC OF
CdS FILM CELL 91H-1

light and dark characteristics are given for the two and three percent cells. Figures 4-7 and 4-8 show the light characteristics of CdS film cells deposited on duPont H-Film. These have nominal efficiencies of one percent.

Detailed analyses of CdS photovoltaic cell characteristics are not possible because no one has yet developed a suitable mathematical (or phenomenological) model of the photovoltaic effect in CdS. However, some empirically determined facts and conclusions have been obtained by workers in this field from examination of a great many i-V characteristics. Several of these are illustrated in the figures. In Figs. 4-4 through 4-6 it is observed that one cell of each pair has a somewhat "softer" reverse charac- teristic and "harder" forward characteristic than the other. Invariably this is the cell with the higher efficiency. This fact has been noted by most workers and has given rise to the notion, "Good solar cells are poor recti- fiers. " In Figs. 4-5 and 4-6 it is seen that the light and dark forward characteristics of a cell are displaced with respect to the voltage axis, the light characteristic being closer to the current axis than the dark character- istic. It is also observed that the displacement is larger for one cell than the other. Nearly always, the more efficient cell exhibits a smaller displace- ment than the less efficient cell. It is believed that the magnitude of this displacement depends on the photoconductivity of the cell and suggests that good solar cells are poor photoconductors. It is also obvious from the figures that the three percent cells have larger short circuit current densities but not necessarily larger open circuit voltages than the two percent cells. The same comparison may be made between the two and one percent cells.

In Figs. 4-4 throgh 4-6, one cell of each pair is from evaporation 87. If one assumes that the eight CdS films from this evaporation were identical, then one must speculate that the differences in the three cells described here were caused by unintentional variations in processing. (The eight 1 inch x 2 inch films were cut from a single 4 inch x 4 inch substrate used in evaporation 87.) Whatever the cause, it is interesting to note that three films, apparently treated identically, result in three cells of one, two and three percent efficiency. It is even more interesting to note that these three cells contradict the maxim that good solar cells are poor rectifiers,

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for the curves show that the three percent cell is a better rectifier than the two percent cell, which in turn is a better rectifier than the one percent cell, particularly in regard to their reverse characteristics and sharpness of the "knees" of the i-V characteristic.

Figures 4-7 and 4-8 show the light characteristics of CdS cells having duPont H-Film substrates. Both cells are nominally one percent efficient, and both cells were processed in the same way. Cells 89H-C and 91H-C represent sub areas of the larger cells are indicated in Table 3-1. The H-Film substrates were 4 inch x 4 inch pieces held in a metal frame. The total area of CdS was about 3.5 inch x 3.5 inch in each case. The films were processed by painting the slurry over four separate areas of each film, each area being approximately 10 or 11 cm^2. Thus, four smaller cells were made on each substrate, each cell having a separate barrier alyer but a common n-type CdS layer. Figures 4-7 and 4-8 show the characteristics of one each of the small cells and the characteristic of the large cell obtained by connecting the four small cells in parallel.

4.6 Mathematical Models

The underlying principles of the photovoltaic effect in silicon solar cells are reasonably well understood; those of CdS not at all. Shockley[4] developed the diffusion theory of the photovoltaic effect in p-n junctions in germanium; Pfann and Scaff[5] applied diffusion theory to silicon p-n junctions; Sah, Noyce and Shockley[6] developed the recombination current theory for silicon which pretty well explained the observed characteristics of silicon solar cells, and a better understanding of silicon was achieved by Shockley and Queisser[7] and Queisser;[8] Anderson[9] studied the photovoltaic effect in Ge-GaAs heterojunctions, and Cusano[10] has studied CdTe p-n junctions; but no one has succeeded in a reasonable analysis of the Cu-CdS barrier layer. Lacking a theory, one must fall back on the purely phenomenological approach taken by Prince,[11] Loferski,[12] Rappaport,[13] and Wolf and Rauschenbach.[14]

These papers relate to silicon and consider a solar cell as being adequately represented by an equivalent circuit consisting of a constant current generator, i_L, shunted by an ideal diode, i_j, which in turn is shunted by a resistance R_{SH}. To the output terminals is added a series resistance R_S. The circuit is shown in Fig. 4-9. Prince, Loferski and Rappaport invoke the high shunt resistance and low series resistance of a good silicon p-n junction diode in order to eliminate R_{SH} and R_S from their calculations, resulting in the relationship

$$i = i_L - i_o(e^{\lambda V} - 1) \ , \tag{4-5}$$

where i and V are the measured current and voltage at the output terminals, i_L is the light generated current, i_o is the reverse saturation current of the diode, and $\lambda = -q/AKT$, in which q is the electronic charge, K and T are the Boltzman constant and absolute temperature respectively, and A is a number greater than 2 for silicon, CdS and most other photovoltaic cells. (For ideal silicon p-n junctions, Sah, Noyce and Shockley[6] show that A is exactly 2. Queisser[8] showed how, for real silicon junctions, A could have values between 2 and 3.) Essentially, then, Prince, Loferski and Rappaport considered only ideal solar cells. Wolf and Rauschenback, on the other hand, chose to consider in detail the circuit in Fig. 4-9 for which the current-voltage relationship is

$$i = i_L - i_o(e^{\lambda(V-iR_S)} - 1) + \frac{V - iR_S}{R_{SH}} \ . \tag{4-6}$$

This is transcendental in both variables as well as in the parameter R_S. Wolf and Rauschenbach showed that this still does not represent in detail the real silicon solar cells available in 1961, and developed first, a distributed parameter circuit represented by a complicated rectangular array of p-n junctions and resistances and next, a second order lumped constant circuit which is essentially that of Fig. 4-9 repeated to the right and with the single diodes replaced by pairs of diodes with slightly different characteristics.
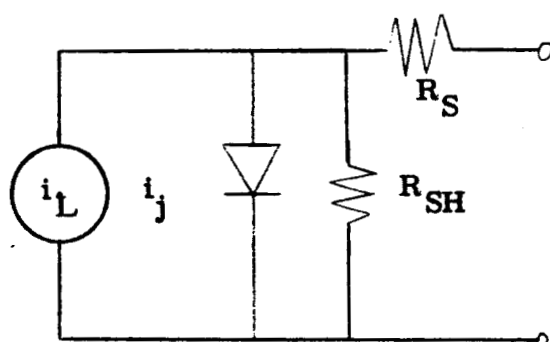
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FIGURE 4-9. EQUIVALENT CIRCUIT OF PHOTOVOLTAIC
SOLAR CELL

Equation (4-6) does represent silicon solar cells quite well and is much easier to handle than the later approximations of Wolf and Rauschenbach. It does not of course, describe CdS cells particularly well; the most notable difference being the CdS reverse characteristic in the 3rd quadrant which suggests a breakdown of some sort. However, the forward characteristic of a CdS cell appears similar to plots of Eq. (4-6), and in order to examine this similarity more carefully, Eq. (4-6) has been solved for various values of the parameters i_L, i_o, λ, R_S and R_{SH}. This has been done on Clevite Corporation's Burroughs 5000 computer. These solutions, in the form of tables of i vs V, are being plotted so a comparison can be made with experimentally determined CdS i-V curves. It is expected that certain of the parameters can be reasonably evaluated so that the computer can than be employed to obtain "best fit" equations for the observed data.

5. ALTERNATE METHODS OF PRODUCING FILMS

Work on high temperature sintering, spraying and pressing CdS films has stopped. In general, sintered and sprayed films formed adherent coatings on glass but not on plastic substrates. These coatings could not be doped sufficiently to lower the resistivity so that large currents could be drawn through the film.

During this quarter efforts have been concentrated (with considerable success) on producing cadmium sulfide mirrors on glass substrates. This is essentially a room temperature process and therefore attractive.

Early work on lead sulfide mirrors was reviewed. A patent issued to W. H. Colbert and W. L. Morgan[15] describes a process for making a lead sulfide mirror and mentions that sulfides of certain other metals, including cadmium, also produce mirrors, but no formulas are listed.

Experiments showed that Colbert's recipe was not directly applicable to the formation of CdS mirrors. Modifications were introduced, and a CdS mirror was successfully deposited on the walls of a test tube and a strip of H-Film contained therein. The film on the glass was very adherent and could not be rubbed off. The deposit on H-Film was fluffy and did not adhere. It appeared that the H-Film was attacked by the reacting solutions.

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Subsequent attempts to deposit lead sulfide mirrors on H-Film were unsuccessful.

The experiments on CdS mirrors are interesting. The principal problems are: 1) achieving slow precipitation of CdS to allow grain growth during the formation of a continuous layer on the substrate, and 2) effective processing of the substrate surface to ensure adherence of the CdS. The question of precipitating dopants along with CdS will be deferred for the present.

For slow precipitation of CdS, complex cadmium compounds are mixed with reagents which release sulfur very slowly. The complex cadmium compounds are formed with ligands, chosen in consideration of their instability constants, which should not exceed the solubility product of CdS (2.6×10^{-29}).

All reactions took place in solutions made alkaline with NH_4OH and had pH values in the range of 10 to 12.

The cadmium salts employed were the acetate, chloride, sulfate and nitrate. The sulfur releasing agents were thiourea, thioacetamide, thiosinamine and cystine.

H-Film substrates were stripes 2 cm x 5 cm and 2.5×10^{-3} cm to 12.5×10^{-3} cm thick. One side of the strips was sandblasted to roughen it.

The experiments generally consisted of forming the complex $\left[Cd(NH_3)_4\right]^{++}$ by adding NH_4OH to the cadmium salt solution until the precipitated $Cd(OH)_2$ was redissolved. Then the solution of the sulfur releasing agent was added in mild excess over the stoichiometric requirement. Approximate concentrations ranged from 0.015M to 0.4M referred to the total volume of the final mixture.

Thioacetamide, CH_3NH_2CS, precipitated CdS almost instantaneously. Cystine, $\left[HO_3C \cdot CH(NH_2)CH_2S\right]$, did not produce CdS at all.

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With thiourea, $(NH_2)_2CS$, and thiosinamine, the formation of CdS was reasonably slow. Presumably, the reactions are:

$$\left[CD(NH_3)_4\right]^{++} + (NH_2)_2CS + 2H_2O \rightarrow \underset{\downarrow}{CdS} + NH_2CN + 2NH_4^+ + 2NH_4OH,$$

and

$$\left[Cd(NH_3)_4\right]^{++} + CH_3CH:CHNH \overset{NH_2}{\diagdown} CS + 2H_2O \rightarrow \underset{\downarrow}{CdS} + 2NH_4^+ +$$

$$+ CH_3CH:CHNHCN + 2NH_4OH .$$
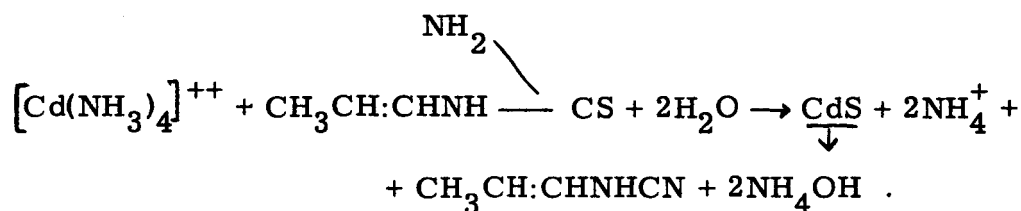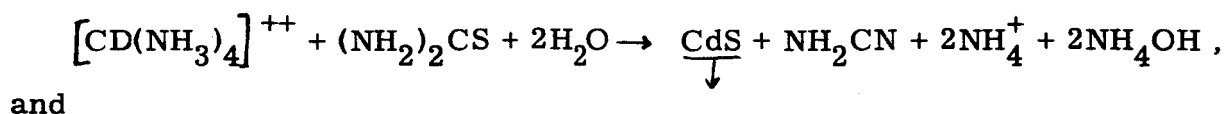
Usually, after 10 or 15 minutes, the mixture became opalescent, then acquired a yellow tint, and after several hours, particle aggregation would begin. The reaction rate could be increased by raising the temperature.

The CdS deposit on H-Film, regardless of pretreatment, and on the walls of plastic test tubes, was at best spotty, very thin and only poorly adherent. On pyrex glass test tubes, and flat substrates, excellent CdS mirrors were obtained from some of the reactions. These films could not be rubbed off the glass, and were thin.

It has been determined that H_2SO_4, NH_4OH and NaOH readily attack H-Film while HNO_3 and HCl do not. Attempts to neutralize the NH_4OH formed in the reactions by controlled addition of acid, or to impede its ionization by adding an ammonium salt, prevented the formation of CdS. This was probably due to suppression of the release of Cd^{++} from $\left[Cd(NH_3)_4\right]^{++}$ by the additives.

The reactions were carried out at room temperature, and the rate of CdS formation was therefore dependent largely on the concentrations of the reagents. Two to 25 hours were required for the formation of the CdS mirror. There seemed to be little difference in the thickness of the films formed. The excess CdS simply settled out as loose sediment.

Of all the Cd salts tried, the sulfate seemed to produce the best mirrors. This will be investigated more thoroughly with emphasis placed on obtaining thicker films on glass substrates.

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6. WORK PLANNED FOR NEXT QUARTER

In the next quarter, emphasis will be placed on fabricating CdS photovoltaic cells on duPont H-Film and improving the characteristics of the cells. An intensive study will be made of cell deterioration, with work continuing on eliminating donor contamination of the barrier as well as the contaminants put down with the slurry. The work on a mathematical model will continued.

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Attention: Georgiana Larrabee, Librarian   1

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Solid-State Electronics Laboratory
Stanford Electronics Laboratories
Stanford University
Stanford, California
Attention: Prof. G. L. Pearson   1

Westinghouse Electric Corporation
Research & Development Laboratories
Churchill Borough, Pennsylvania
Attention: H. C. Chang   1

Westinghouse Electric Corporation
Semiconductor Division
Youngwood, Pennsylvania
Attention: Don Gunther   1